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Analysis and the understanding of fluoride removal mechanisms by an electrocoagulation/flotation (ECF) process

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ABSTRACT

Electrocoagulation is a method of applying direct current to sacrificial electrodes that are submerged in an aqueous solution. Dissolving aluminum (Al^{3+}) is predominant in the acidic condition and aluminum hydroxide has tendency soluble. The defluoridation process was found to be efficient for a pH ranging from 6 to 8. The fluoride removal mechanisms are investigated based on the solution speciation (Al and Al–F complexes) and dried sludge characteristics in the electrocoagulator. The XRD analysis of the composition of the dried sludge shows the formation of $Al(OH)_{3-x}F_x$ and provides confirmation for the main mechanism for fluoride removal. The mechanism of the fluoride removal was confirmed to be not only the competitive adsorption between OH^- and F^- but also the formation of solid cryolite in pH range of 5–8.

1. Introduction

Inorganic constituents, which may be presented in natural waters or in contaminated source waters, are found to become a major public health problem in drinking water. The presence of fluoride, as an inorganic ion, is serious more than limits in drinking water and it is a public health problem. The fluoride removal in drinking water and in wastewater has been the subject of many publications and studies that have progressively developed the aspects of toxicity on man and on the environment. When the fluoride concentration is increased to more than 4 mg L^{-1} in drinking water, fluorosis deformity at hips, knees and other joints are seen in skeletal fluorosis [1]. However, long-term use of low fluoride concentration (less than 0.5 mg L^{-1}) is the cause of dental caries [2]. Because of the public health significance of high fluorides consumption in drinking water, defluoridation is important. The maximum acceptable concentration of fluoride in water is 1.5 mg L^{-1} [3].

Fluoride pollution occurs through two different sources including natural and anthrogenic sources. In groundwater sources, the natural concentration of fluoride depends on the geological, chemical and physical characteristics of the aquifer, as concentrations of up to 38.5 mg L^{-1} have been reported in India. Some of the water supplies can be polluted by discharging of industrial wastewater without any fluoride treatment, including glass manufacturing industries [4] and semiconductor industries [5] in the natural environment. Several methods were globally studied for defluoridation of water, such as: adsorption [6–8], chemical precipitation [9–15], electrodialysis [13], and electrochemical methods [16,17]. These methods can be divided in two categories as precipitation and sorption methods [17]. Lime addition is the most common technology of the first group and is used for high fluoride concentration. Lime is used to form CaF_2 precipitate and reduce the fluoride concentration. In laboratory experiments, a two-column limestone reactor has been designed to reduce fluoride concentrations from 109 mg L^{-1} to 4 mg L^{-1} [12]. The second groups (sorption methods) need regular column regeneration and not cost-effective to treat wastewaters with fluoride concentration greater than 10 mg L^{-1} [12]. Lime is the cheapest chemical used for the defluoridation of wastewater, however, it is impossible to reduce the fluoride concentration to 1 mg L^{-1} using only lime [6] .Toyoda and Taira [5] proposed a new method for treating high fluoride concentration (100 mg L^{-1}) to reduce sludge and running costs at two stages, e.g., the formation of CaF_2 by addition of Ca salt such as $Ca(OH)_2$ and the adsorption of the residual fluorine by $Al(OH)_3$ by addition of an Al salt. In the 1st step of the conventional treatment system, fluoride concentration was decreased from 100 to 20 mg L^{-1}, since it can be decreased to 2.5 mg L^{-1} by addition of Al salt and neutralizing in the 2nd step. Using chemical coagulants for precipitation is one of the most essential methods in conventional water and wastewater treatment. However, the generation of large volumes of sludge, the hazardous waste categorization of metal hydroxides, and high costs associated with chemical treatments have made chemical

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